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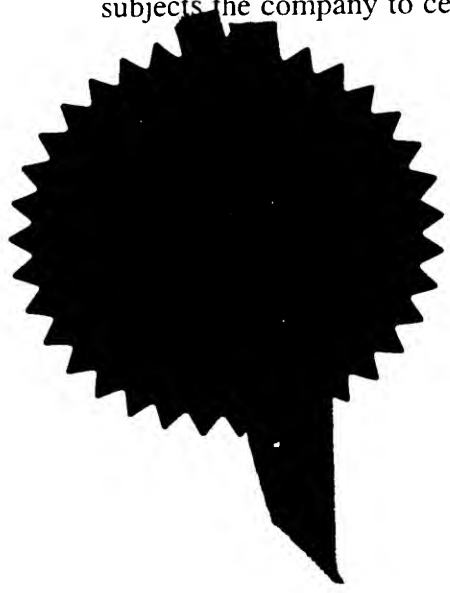
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Signed

*Andrews*

Dated

11 October 2000



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The Patent Office

Cardiff Road  
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Gwent NP9 1RH

1. Your reference

MPD309/III/GB/RGMS

2. Patent application number

(The Patent Office will fill in this part)

07 JUL 1996

9923593.9

3. Full name, address and postcode of the or of each applicant (underline all surnames)

ALBRIGHT & WILSON UK LIMITED  
210-222 HAGLEY ROAD WEST  
OLDBURY  
WEST MIDLANDS B68 0NN

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

ENGLAND

4. Title of the invention

STRUCTURED SURFACTANT SYSTEMS

5. Name of your agent (if you have one)

R G M SAVIDGE

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

ALBRIGHT & WILSON UK LIMITED  
PATENTS DEPARTMENT  
210-222 HAGLEY ROAD WEST  
OLDBURY  
WEST MIDLANDS B68 0NN

Patents ADP number (if you know it)

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number  
(if you know it)

Date of filing  
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing  
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

NO

- a) any applicant named in part 3 is not an inventor, or
  - b) there is an inventor who is not named as an applicant, or
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- See note (d))

# Patents Form 1/77

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Claim(s)	NONE
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Priority documents	NONE
Translations of priority documents	NONE
Statement of inventorship and right to grant of a patent (Patents Form 7/77)	NONE
Request for preliminary examination and search (Patents Form 9/77)	NONE
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Any other documents (please specify)	NONE

11. I/We request the grant of a patent on the basis of this application.

Signature  Date 06/10/99  
R G M SAVIDGE By Power of Attorney

Name and daytime telephone number of  
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MPD309/iii

PATENTS ACT 1977

PRELIMINARY SPECIFICATION  
(Description)

ALBRIGHT & WILSON UK LIMITED

Applicant :

ALBRIGHT & WILSON UK LIMITED

Inventors :

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JOHN HAWKINS

### STRUCTURED SURFACTANT SYSTEMS

The invention relates to a novel type of structured surfactant system. The novel system is especially suitable for suspending pearlising concentrates for incorporation into liquid formulations such as shampoos and toiletries to impart a nacreous iridescence which is attractive to consumers, and can mask inhomogeneities in the formulations.

Structured surfactant systems have the ability to suspend solid particles indefinitely in a pourable liquid. While the system is at rest it behaves like a solid gel holding the particles immobile, but the shear forces associated with pouring break the structure causing the suspension to flow like a mobile liquid. Known structured surfactant systems are either opaque interspersions of a surfactant mesophase with an aqueous phase or cloudy opalescent lamellar phases.

Pearlisers typically comprise small, thin, transparent platelet crystals which can be suspended in a parallel configuration. When so suspended light falling on the crystals undergoes complex multiple reflections within the substrate similar to those which occur in a pearl and giving rise to similar optical interference effects.

Natural pearls comprise alternate layers of calcium carbonate and protein. Artificial pearlisers are made by preparing thin, transparent, platelet shaped crystals from salts of lead, zinc, mercury and bismuth (e.g. bismuth oxychloride), titanium oxide and various fatty acid derivatives such as magnesium stearate, coconut monoethanolamide, ethylene glycol distearate and ethylene glycol monostearate. Fish scale extracts are too expensive and the inorganic pearlisers are either too toxic for general use in toiletries e.g. lead, mercury, or relatively ineffective e.g. bismuth. The fatty acid derivatives are therefore now the most widely used pearlisers. In addition to the chemical nature and physical form of the pearliser the manner in which it is suspended has an important effect on its visual impact. Difficulty is sometimes encountered obtaining the desired effect when incorporating pearlisers into aqueous formulations.

Conventional fatty acid derived pearlisers are supplied as solids which are usually added to a heated formulation above their melting point and recrystallised in situ. The conditions of crystallisation and especially the amount and nature of the agitation applied must be carefully

controlled in order to obtain an acceptable result. This makes it difficult to obtain consistent effects and renders solid pearlisers inconvenient to use.

Attempts have been made to prepare liquid concentrates or suspensions which can be added directly to shampoo formulations without heating. While more convenient for the user, such concentrates face the manufacturer with problems of obtaining a high and consistent pearl effect, similar to those which confront the user of conventional solid pearlisers. Difficulty is also encountered in maintaining the particles in stable suspension and preventing sedimentation.

We have now discovered that certain surfactants having a high solubility parameter such as alkyl ether sulphates at concentrations just below those which normally give rise to liquid crystal phases, e.g. typically 18 to 30% by weight, will form mobile, clear protolamellar  $L_1$  phases in the presence of sufficiently high concentrations of electrolyte. These mobile phases are clear and optically isotropic and are typically less viscous than normal G-phases but have the capacity to form stable suspensions of solids such as pearlisers.

The protolamellar phase is apparently unique in being clear and optically isotropic while exhibiting the suspending properties of a structured surfactant system. We believe that protolamellar phases comprise oblate (disc) micelles which can adopt a parallel lamellar-like orientation. They may typically be identified by placing them between crossed polarisers and applying stress, e.g. by tapping or shaking, when flashes are observed as the system transiently forms an anisotropic structure.

When pearlisers are suspended in a protolamellar phase the structure promotes the parallel orientation of the pearlisng crystals which maximises the pearlescent effect, even in the absence of agitation. The concentrates are readily pourable but do not generally tend to separate on standing, and can easily be added to shampoo formulations, without heating, to give high and consistent pearlisng effects.

The compositions are particularly useful because the preferred surfactant, alkyl ether sulphate, is the most commonly used ingredient of shampoo formulations on account of its skin mildness and comparatively good foaming properties. Thus the concentrate can be included in shampoo formulations without introducing anything extraneous to the formulator's requirements. The optical clarity of the protolamellar phase permits the pearlescent effect to be clearly observed without being obscured by surfactant structures as occurs when any hitherto known structured surfactant is used.

When alkyl ether sulphates or similar water soluble surfactants are dissolved in water at relatively low concentrations they form a clear micellar solution (L1 phase) with the surfactant molecules arranged in spherical clusters (micelles). As the concentration is increased the micelles form rods (prolate micelles) of increasing length and the viscosity increases. Further increases of concentration are accompanied by an anomalous fall in viscosity as the rods become aligned. The aligned rod micellar or protohexagonal system is optically clear, pourable and may exhibit a hexagonal symmetry when examined by small angle x-ray diffraction (SAX), giving a first order peak of momentum transfer vector  $Q_1$ , and, if the system is sufficiently well defined, one or more, progressively smaller, higher order peaks at  $Q$  values in the ratios:

$$Q_2 = Q_1 ; \quad Q_3 = 2Q_1$$

These ratios are diagnostic of hexagonal symmetry.

When the concentration is further increased the length of rods increases indefinitely and the immobile M phase is formed. This is typically observed at concentrations of about 30% by weight surfactant. The M phase also shows SAX peaks in the ratios characteristic of hexagonal symmetry. It typically resembles a curdy or gelatinous solid or very viscous mucous like fluid. The normal lamellar, or G phase is typically formed when the concentration of the alkyl ether sulphate is increased to about 55 to 60% by weight. It is a transparent, pourable, fluid, which may be solidified by the addition of a crosslinking agent. The G phase is revealed by SAX peaks with the characteristic ratios  $Q_2 = 2Q_1$  ;  $Q_3 = 3Q_1$ . The peaks indicate structure with a repeat spacing ( $d =$ ) of about 3 to 5 nm. Although the normal G-phase has a yield point and is relatively mobile compared with the M-phase it is not generally suitable for use as a suspending medium for solids, since the presence of appreciable amounts of suspended solid renders it unpourable. The high surfactant concentration is a further disadvantage.

However, when sufficient electrolyte is added to a micellar or protohexagonal solution of alkyl ether sulphate there appears to be a change from prolate to oblate micelles and corresponding shift from hexagonal to lamellar symmetry with a  $d$  spacing typically in the range 6 to 15 nm. Such phases are able to suspend solids and remain readily pourable.



Our invention therefore provides a suspension of solid particles in an aqueous surfactant, wherein said surfactant is in a protolamellar phase. In a preferred embodiment the suspended particles comprise particles of a pearliser.

The surfactant is preferably a C<sub>12-14</sub> alkyl or alkoxyl 1 to 20 mole ethoxy sulphate, more usually a C<sub>12-14</sub> alkyl ethoxy sulphate and preferable an alkyl 2 to 10 mole ethoxy sulphate. Alternatively the ether sulphate may be a mixed propoxy ethoxy sulphate or alkyl glyceryl or alkyl glyceryl polyethoxy sulphate. The cation of the alkyl ether sulphate is preferably sodium but may be potassium, lithium, ammonium, or an amine such as an alkanolamine.

The surfactant may comprise minor amounts of other surfactant, especially anionic, cationic or amphoteric surfactants, for example alkyl sulphates, alkyl benzene sulphonates, paraffin sulphonates, olefin sulphonates, alkyl sulpho-succinates, soaps, taurides, isethionates, alkyl ethoxylates, fatty acid ethoxylates, alkyl glyceryl ethoxylates, alkyl carbohydrate ethoxylates, amine oxides or betaines. However it is generally preferred to use essentially only alkyl ether sulphate.

The electrolyte is preferably sodium chloride, but could be, for example, sodium carbonate, sodium citrate, sodium tripolyphosphate, sodium hydroxide or any other salt or base which tends to salt alkyl ether sulphates out of solution. The cation of the electrolyte may be sodium, potassium lithium, ammonium or, less preferably, an amine such as alkanolamine or a mixture of any of the foregoing.

ingredients the formulator, to whom the concentrate is to be supplied, wishes to include in the end formulations, or can tolerate.

Generally the surfactant is used in a total concentration which, in the absence of electrolyte, would correspond to a clear L<sub>1</sub> phase or protohexagonal phase. Typically the electrolyte-free surfactant/water system would be unstructured or would exhibit hexagonal symmetry under SAX. The concentrations required vary with different surfactants but are generally in the range 18 to 35% based on the total weight of surfactant and water, more usually 20 to 32% e.g. 25 to 30%.

The amount of electrolyte is sufficient to convert the ether sulphate/water mixture from an optically isotropic, clear, L<sub>1</sub> phase or a protohexagonal phase exhibiting hexagonal symmetry to a clear, protolamellar phase, typically exhibiting lamellar symmetry, under SAX. The

amount required will depend on the nature and concentration of the surfactant. Typically it will be between 5 and 25% based on the total weight of surfactant, electrolyte and water, more usually 6 to 20% e.g. 8 to 18%.

The pearlisers may be of any of these principles, however, they are preferably organic rather than inorganic pearlisers but is preferably a fatty acid derivative especially a mixture of ethylene glycol distearate and ethylene glycol monostearate.

The pearlisers may be dispersed in the aqueous structure surfactant system e.g. by gently stirring, but in the case of the fatty acid derivatives are preferably prepared in situ by heating above their melting point (e.g. temperatures between 65 and 80°C) dispersing the pearlisers in the structured surfactant system, preferably with sufficient stirring to form droplets of from 0.5 to 20 microns, e.g. 1 to 10 microns, and cooling to ambient temperature. Preferably cooling is relatively slow e.g. the mixture is allowed to cool naturally. The amount of pearlisers can be varied considerably, the main constraint on the upper limit being the viscosity.

The amount of pearlisers should not be so high as to render the product unpourable, or unacceptably viscous. We prefer on economic grounds that the pearlisers are present in amounts greater than suspending surfactant. Generally pearlisers may be present in amounts ranging from 5% up to about 50% e.g. 10 to 45% of the total weight of the mixture.

Ancillary ingredients suitable for inclusion in toiletries could in principle be included, but are not included in the present formulation. However, it is generally desirable to include small amounts of preservative, such as formaldehyde or other microbiocidally and/or fungicidally active material.

The novel structured, protolamellar phase may be used to suspend solids other than pearlisers, e.g. builders such as zeolite or phosphate for use in detergent. They may also suspend bentonite or calcite. They may suspend rock cuttings and/or weighting agents for use in drilling muds. They can suspend agricultural pesticides or dyes or pigments, dicalcium phosphate for use in toothpaste or ammonium polyphosphate for fire retardants.

The invention is illustrated by the following examples -

	<u>WT%</u>
Sodium C <sub>12-14</sub> alkyl 2-mole ethoxysulphate	21.00
Ethylene glycol distearate (pearliser)	20.00
Sodium Chloride	7.00
Water	balance

The alkyl ether sulphate, as a 70% by weight active G-phase, was diluted with the required amount of water and the sodium chloride was dissolved to provide a clear, homogeneous, pourable, optically isotropic, liquid phase. The composition when viewed through polarising microscope appeared uniformly dark. However, when the cover slip was gently tapped, bright flashes of light were observed, indicating the presence of a protolamellar phase.

The composition was heated to 80°C and the pearliser was melted and added to the composition with vigorous stirring. The composition was allowed to cool while gentle agitation was maintained. A stable suspension with an intense pearly appearance of exceptional quality was obtained.

The composition was reheated to 80°C to melt the pearliser and allowed to cool without further agitation. A stable, high quality pearl effect was obtained. This illustrates the fact that conditions of stirring and cooling are not critical in order to obtain a good pearling

